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X-ray study of Nd:YAG on (111)-oriented Si obtained by pulsed laser deposition

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ABSTRACT

Yttrium aluminium garnet ($\text{Y}_3\text{Al}_5\text{O}_{12}$) thin films doped with neodymium have been prepared by Pulsed Laser Deposition (PLD) method on (111)-oriented Si substrates. The substrate was heated up to temperature in the range between 200 and 600 °C. Obtained films were then characterised both by X-ray diffraction (XRD) method using Siemens D5000 diffractometer and radioluminescence spectroscopy.

Keywords: Yttrium aluminium garnet, pulsed laser deposition, XRD, radioluminescence.

1. INTRODUCTION

Pulsed Laser Deposition (PLD) is highly flexible technique for fabrication of thin films. We study Nd:YAG thin films due to their technological applications in optoelectronic systems. Several deposition techniques, such as liquid-phase epitaxy, have been employed to deposit Nd:YAG thin films, but only few publications on pulsed laser deposition of YAG have been reported. These films have been singled out for studies because of their significance in optoelectronic systems. The effects of the substrate temperature on the film orientation and composition and annealing in air have been investigated by the X-ray diffraction (XRD). The quality of the film was also assessed by measuring their photo- and radioluminescence spectra. The emission lines due to the $4f^6$ intra-configuration transitions of Nd^{3+} ions have been identified and analysed. The films produced by PLD are non-uniform in thickness and the thickness profile is determined by the angular distribution of the mass flux of evaporated material¹. The quality of film strongly depends on substrate temperature². We have also investigated effect of electric field applied between target and substrate.

2. EXPERIMENTAL DETAILS

The films were deposited using the standard experimental set-up³. The stainless steel vacuum chamber was evacuated to a pressure of $1 \cdot 10^{-5}$ mbar. Powders of Y_2O_3 , Al_2O_3 and Nd_2O_3 (Aldrich Chem. Co.) were used as the target for deposition process. A XeCl excimer laser beam ($\lambda = 308$ nm, $\tau = 20$ ns, repetition: 15 Hz, fluence: 8 J/cm^2) was used for ablation. Target was rotating about 18 rpm. The distance between Si substrate and targets was about 6 cm.

3. RESULTS

3.1. XRD spectra

Thin YAG films have been measured by means of X-rays, using D5000 Siemens diffractometer. In that manner some characteristic structural features of the crystals could be determined but as a rule it was not possible to establish relation concerning those features and parameters of the deposition process. The problem is discussed later.

The main goal of performed XRD analysis was to clarify the two major questions as to structural properties of the deposits. At first, long-time stability of the films was investigated, i.e. it was considered whether structural features of the films do alter under ordinary heat treatment, namely annealing. In fact, long-time stability plays the key role from the point of view of fabrication of many various optoelectronic devices, especially high-power ones. Apart from that, XRD measurements have provided data both on the crystal structure of the films and deposit-substrate interlayer. Those could characterise the PLAD method itself, for example as a result of comparison all the spectra and pointing at their common features.

In the case of SAMPLE A four XRD spectra have been recorded, as presented in Fig.1. Those spectra differ both in sample orientation (on the right, sample was turned within 30° with respect to its normal), and in annealing. Detailed analysis provides the following observations:

- (1) thin YAG film seems to be of good quality indicating by a sharp and distinctive peak at $2\theta = 31.45^\circ$ (indexed as YAG(411) or YAG(330)),

- (2) the above results in the lattice constant $a = 12.055 \text{ \AA}$ which proves that there are tensile stresses induced in the layer,
- (3) since the three other peaks permanently appear in the XRD pattern and their intensities do not depend on the sample orientation it implies that some contribution of disordered phase (existing probably at the YAG-substrate interlayer) must be taking into account,
- (4) after annealing XRD signal is no longer detected which may be caused by re-evaporation of the components attesting to poor long-time stability of the deposited film.

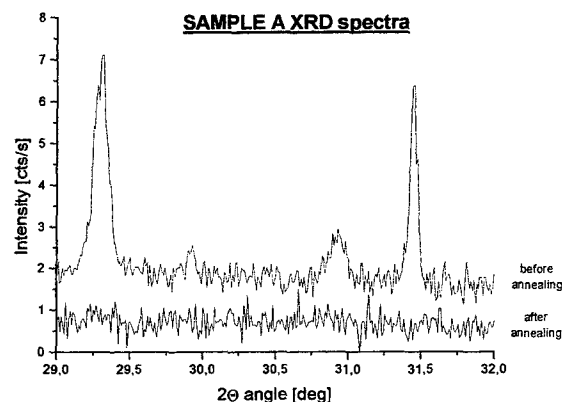
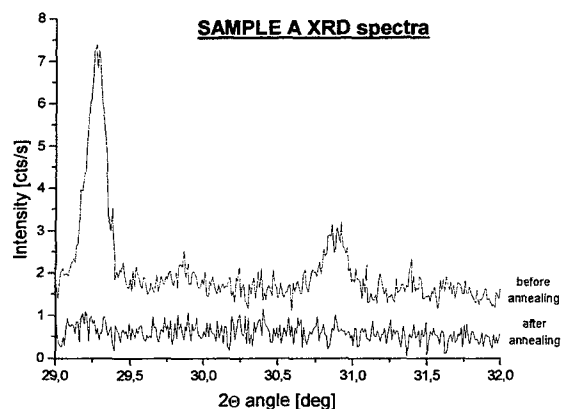


Fig.1. XRD spectra recorded at two different SAMPLE A orientations with respect to its normal: (left) $\phi = 0^\circ$, (right) $\phi = 30^\circ$. Common features are ascribed to strongly disordered structure of the interlayer while sharp peak centred at $2\Theta = 31.45^\circ$ is indexed as YAG(411) or YAG(330). Disappearing of the signal after annealing is observed as well.

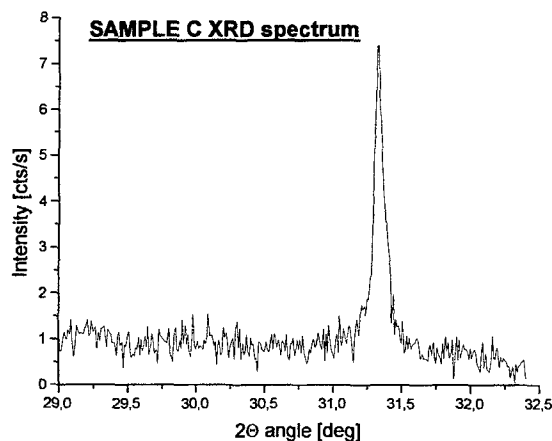
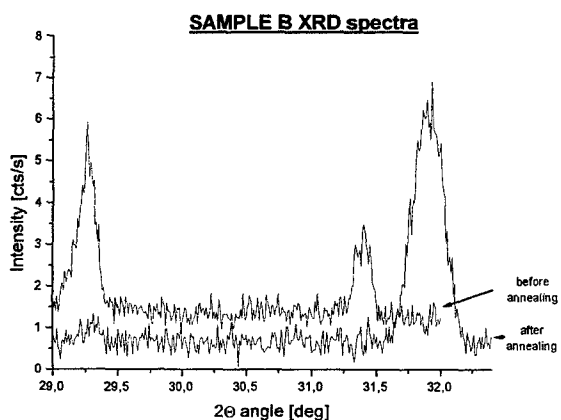


Fig.2. SAMPLE B XRD spectra measured before and after annealing. It appears that under heat treatment peak centred at $2\Theta = 31.40^\circ$ both shifts from its un-annealed position within about 0.5° and increases in its intensity.

Fig.3. SAMPLE C XRD spectrum coming from un-annealed film in which the only one distinctive feature at $2\Theta = 31.32^\circ$ is observed.

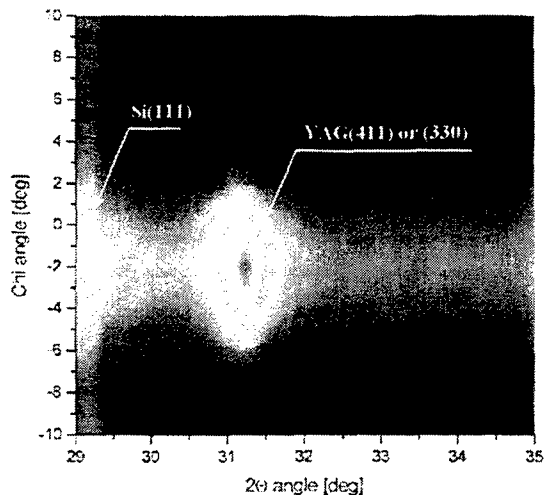


Fig.4. 2-dimensional projection of SAMPLE D XRD pattern plotted both against 2θ as well as χ co-ordinates. Peak shown partly in the left of the figure is Si(111) while that at the centre is YAG(411) or YAG(330). On account of high intensity of the latter (being of the order of magnitude greater than those previously presented) one can state that ordering of the deposit occurs rather than stress-relief behaviour.

Fig.4 shows 2-dimensional projection of XRD pattern of SAMPLE D measured by simultaneous scanning both in 2θ as well as χ co-ordinates. Observed peak is centred at $2\theta = 31.23^\circ$ and $\chi = -2^\circ$. Detailed analysis reveals that Si(111) peak is also centred at $\chi = -2^\circ$ (see on the left of Fig.4). According to that one can conclude that YAG(411) (YAG(330)) planes are parallel to those of Si(111) and therefore heteroepitaxial growth of YAG on Si substrate occurs. Furthermore, predominant orientation of the film is (411) or (330). The highest intensity of the peak (not shown in such projection), being of the order of 200 cts/s, results undoubtedly from considerable thickness of the film. It is not, however, clear whether structure of such thickness is not nearly stress-free (determined lattice constant a is 12.139 Å and is the highest measured). Substantially high FWHM of the peak (about 0.5° and near 3° along 2θ and χ axis, respectively) also points at strong stresses induced in the layer. On the other hand, no signal of the disordered phase is recorded which may be caused by aforementioned heteroepitaxial matching of the two structures.

3.2. Radioluminescence spectra

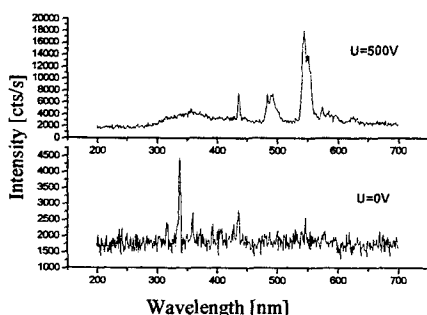


Fig.5. Radioluminescence spectra of Nd:YAG thin films for different voltage between target and substrate. The substrate temperature was 520°C .

In contrast to the previous sample, structure of SAMPLE B seems to be substantially different. It also exhibits quite sharp peak at $2\theta = 31.40^\circ$ indexed as YAG(411) or YAG(330) (before annealing), but another similarities are no further maintained. According to Fig. 2, annealed film reveals very distinctive feature at $2\theta = 31.90^\circ$ so that shift of about 0.5° is observed (with respect to un-annealed sample). In addition, intensity of the peak increases by a factor of 2 after annealing. Assuming that under heat treatment stress-relief behaviour occurs one can expect diminishing of induced stresses. However, observed shift is as high as 0.5° , which indicates that tensile stresses turns into compressive ones (lattice constant a changes from 12.076 to 11.892 Å before and after annealing, respectively). All those suggest that ordering of the growing crystal occurs rather than its stress-relief behaviour. In fact, thermally excited atoms are able to move slightly from their previous positions into new ones in order to decrease energy of the crystal (minimisation of the energy). As a result, rearrangement of the structure is observed.

In the case of SAMPLE C there appears only one diffraction peak in the whole range investigated, centred at $2\theta = 31.33^\circ$ and indexed as YAG(411) or YAG(330). This gives lattice constant as high as 12.103 Å (see Fig.3). Unfortunately, similar heat treatment of the sample was not carried out so that comparison of the results with those previously obtained was not possible.

In Fig.5 we present radioluminescence spectra of Nd:YAG thin films for two different voltages between the target and the substrate. The spectrum of the film obtained under the voltage of 500 V clearly shows sharp lines that are due to $4f^2$ intraconfigurational transitions of the Nd^{3+} ion. The lines, at about 430, 480 and 550 nm, correspond to transitions that originate at lower lying levels. There is, as expected, no $d-f$ emission but there are also some missing $f-f$ emission lines that originate at some higher lying levels of the $4f^2$ configuration and are usually visible in the Nd:YAG monocrystal radioluminescence spectra. The spectrum reveals also some hardly visible broad band emission at 300 to 400 nm that could represent the so-called "host" emission, characteristic of the undoped YAG. Interestingly the second spectrum, of the Nd:YAG film obtained under no voltage, shows no emission at all.

In Fig.6 we show a photoluminescence spectrum of the Nd:YAG thin film that was annealed in air for 1 hour at 800°C . The

exciting light wavelength was set at 186 nm. The spectrum reveals a broad band that resembles the "host" emission band. There also is a rich structure imposed on the broad band that could be due to Nd^{3+} $f-f$ emission lines observed in some Nd:YAG monocrystals.

Finally, in Fig.7 we present radioluminescence spectra obtained at three different spots of the same Nd:YAG film the spectrum of which was shown in Fig.5 (500 V). The differences between the spectra are quite likely indicative of the quality of the local crystalline environment and/or statistical variations of the local Nd concentration.

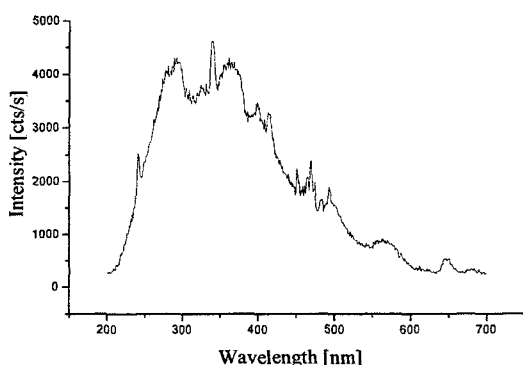


Fig.6. Photoluminescence spectrum of Nd:YAG. The sample was annealed in air for 1 hour in 800 °C.

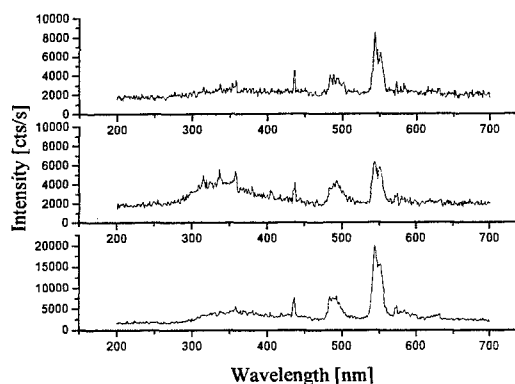


Fig.7. Radioluminescence spectra coming from the three different regions of the same film.

4. CONCLUSIONS

In the present work, the Nd:YAG thin films on (111)-oriented Si substrates have been obtained by two techniques: PLD method with electric field between target and substrate, and PLD method with annealing in air. Those have been proved to possess some interesting structural and electronic properties that have been determined using X-ray diffractometry supported by radioluminescence spectroscopy. XRD patterns reveal that performed annealing may lead either to re-evaporation of the film components or to ordering of atom arrangement in a deposited crystal. In spite of that, however, stress-relief behaviour does not occur. The absence of some $f-f$ emission lines in radioluminescence of Nd:YAG films that are present in the spectrum of Nd:YAG monocrystals calls for further studies. This observation could potentially provide new and interesting information about (quenching, energy transfer) processes that are of importance in the films. It is also interesting to note that radioluminescence can potentially provide a convenient measure of the film quality.

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